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(71) Applicant (for all designated States except US): H. B. FULLER  
LICENSING & FINANCING, INC. [US/US]; 1200 Willow  
Lake Boulevard, St. Paul, MN 55110-5132 (US).

(72) Inventor; and

(75) Inventor/Applicant (for US only): JANSSEN, Annegret  
[DE/DE]; Mühlenkamp 8, D-21337 Lüneburg (DE).

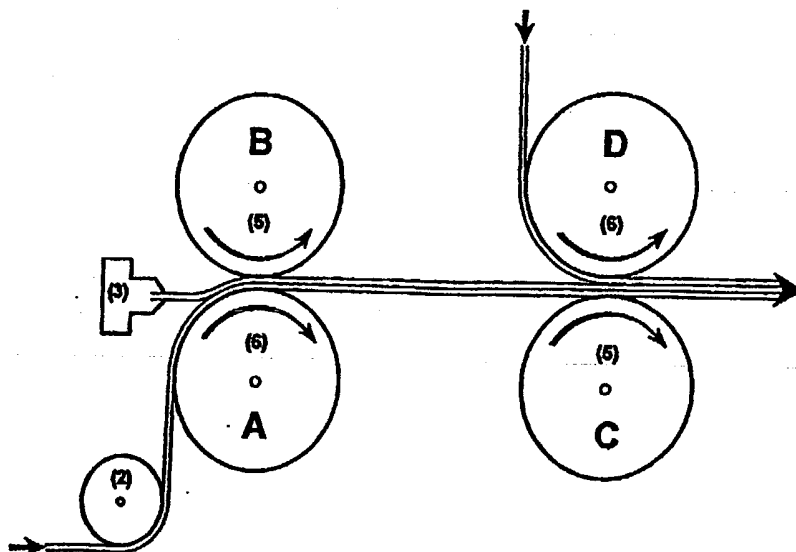
(74) Agent: MAIWALD, Walter, Maiwald GmbH, Poccistrasse 11,  
D-80336 München (DE).

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(57) Abstract

This invention relates to a non-contact coating method for producing a substantially continuous coating and articles constructed therefrom. This invention further relates to a non-contact slot coating method for producing a variety of coatings and laminations. This invention particularly relates to a method of coating a non-porous substrate including film, foil and paper with a molten thermoplastic composition which reduces streaking caused by particles such as impurities, fillers and superabsorbent polymers. This invention further relates to thermoplastic compositions useful in the present non-contact coating method.

- 5 mechanical and thermal stresses on the coated substrates, especially since the slot nozzle is heated during coating. Therefore, very sensitive substrates such as plastic films can not always be coated with a hot melt from a slot nozzle in a customary manner without damaging the substrate.
- 10 Further, the high coating weights of this prior art lead to increased stiffness of the coated substrate.
- WO 96/25902, published Aug. 29, 1996, assigned to the H.B. Fuller Co. in St. Paul, MN teaches a method of coating
- 15 wherein certain thermoplastic compositions are thermally made flowable and released from a coating device as a continuous coating without contact between the coating device and substrate being coated.
- 20 The present invention resides in specific adaptations to this novel coating method for use in a variety of other applications involving coatings on nonporous materials and coatings on porous materials. One type of such application is coatings on nonporous materials such as films.
- 25 Thermoplastic compositions often contain unmelted particles either in the form of impurities such as contaminants and char or alternatively in the form of a particulate ingredient such as filler and additives. When these particles are of appreciable size and/or the slot nozzle has
- 30 a relatively small gap, the particles tend to accumulate in the coating device interfering with the deposition of the coating. The particles block the passage of thermoplastic material causing a corresponding striation or streak to form on the substrate being coated. This problem is particularly
- 35 prevalent in the formation of very thin coatings particularly when the optical quality is of importance such as for high quality graphic art applications, especially where films have to be coated. Accordingly, industry would find advantage in a coating method which rectifies these

5 problems.

It is therefore an important object of this invention to provide a new coating method especially suited for coatings on films, foils, paper and other such materials, which makes  
10 it possible to avoid streaking and striation problems, especially at very low coating weights.

It is another important object of the invention to provide a coating method which permits laminations and coatings to be  
15 carried out „inline“ or „offline“, using thin films, metallized foils, heat-sensitive materials and other sensitive substrates at reduced risk of obtaining faulty or flawed products.

20 It is yet another important object of the invention to make film-to-film and film-to-foil laminations available which do not require the use of reactive adhesives.

Another object of the invention is to provide improved  
25 coating methods for coating thermoplastic compositions, especially hot melt adhesives, onto porous substrates such as textiles.

These and other objects and advantages of the invention will  
30 become more apparent from the following discussion.

#### Summary of the invention

35 The present invention is a method of coating a substrate with a thermoplastic composition employing a non-contact coating method and articles constructed therefrom. The method produces a substantially continuous coating. The method is useful for a variety of adhesive and coating

- 5 for coating and mutually bonding thermally sensitive substrates.

The thermoplastic composition preferably exhibits certain rheological characteristics such that the complex viscosity  
10 at high shear rates (1,000 rad/sec) is less than about 500 poise and the complex viscosity at low shear rates (1 rad/sec) is less than about 1,000 poise at the coating temperature. Some neat thermoplastic resins are suitable for the method of the present invention provided the  
15 uncompounded materials are sufficiently low enough in viscosity. However, compounded hot melt adhesives are preferred due to the ability to independently control the viscoelastic properties, open time, etc. Compounded hot melt are also advantageous to insure adequate adhesion to  
20 the carrier substrate or for delayed detackification of the coating after adherence to the substrate.

The resulting coating produced from said method is useful for a variety of applications wherein a consistent nonporous  
25 substantially continuous coating is desired. Coating weights of less than about 50-60 g/m<sup>2</sup> are preferred and even more preferred are coating weights of less than about 30 g/m<sup>2</sup> of the thermoplastic composition due to reduce expenditure and improved tactile quality of coated  
30 substrates. Coating weights of less than 10g/m<sup>2</sup> can in many cases be achieved.

The resulting coating is preferable for producing laminations to paper or cardboard, especially to printed  
35 paper. The coating method is particularly advantageous for manufacturing as it employs fewer production steps than prior art coating methods. Improving productivity as well as reducing the coating weight mass per area results in coatings and corresponding articles that are less expensive

5 than prior art.

The coating methods are however not restricted to applications involving nonporous substrates. The inventive coatings can also be used on porous substrates. Herein,  
10 various aspects of the invention can be employed, including a method where the thermoplastic composition is released from a coating device with the distance between the coating device and the substrate greater than 20 mm, and including the method of nipping a hot melt adhesive preformed film to  
15 a porous substrate by means of a release coated roller in direct contact with the adhesive film.

Articles as described herein comprise articles which have at least one first layer wherein said first layer is a  
20 nonporous substrate and at least one second layer wherein said second layer is a coating or adhesive layer produced from the coating methods described above.

## 25 Brief description of the drawings

Figs. 1-10 depict some of the preferred embodiments of the method of the present invention wherein a substantially continuous thermoplastic coating is formed and adhered to a  
30 substrate.

More specifically, Fig. 1A shows the basic structure of a coating and laminating machine useful for operating the invention;  
35

Figs. 1B and 1C show the basic structures of similar such machines;

Figs. 2-4 illustrate inventive laminations at different

5 positions of the coating device;

Figs. 5A and B illustrate a lamination and a transfer-coating method according to the invention;

10 Figs. 6-10 illustrate laminations, including adhesive reactivation laminations, in accordance with the invention.

#### Detailed description of the invention

15

In the method of the present invention, a molten thermoplastic composition, such as a hot melt adhesive, preferably substantially air-free, is initially provided in the form of a substantially continuous, nonporous "film" which is only later contacted with a substrate, a transfer roller or some other kind of support. Generally, the composition is released from a coating or release device in such a way that it exits the device as a substantially continuous film. A typical coating device is a slot nozzle, as it has previously been used for coating in direct contact with substrates. Thus, hot melt coating devices which are already known can be employed in accordance with the method of the present invention in that the slot nozzle is lifted off the substrate and is adjusted to have a suitable distance from the substrate.

As the flowable molten adhesive or thermoplastic composition exits the coating device, it does not contact the substrate, but rather travels for a distance as a continuous film suspended between the coating device and the substrate. The coating device may be initially contacted to the substrate in order to anchor or adhere the thermoplastic composition to the substrate provided that substrate will not be thermally or mechanically damaged by the contact with the

5 coating device. Alternatively, the thermoplastic composition exits through the nozzle as a substantially continuous film and descends until contacting the substrate. The leading edge of the advancing substantially continuous film of thermoplastic composition adheres or anchors to the  
10 substrate upon contact with the substrate. In the case of heat sensitive materials, it is advantageous to advance the substrate by means of the drive rolls prior to contacting the thermoplastic composition to the substrate to avoid a build up of molten material which will melt through the  
15 substrate.

Machinery suitable for operating the inventive methods is shown schematically in Figures 1A, 1B and 1C. Figures 1A and 1B show an embodiment where a thermoplastic composition  
20 is released from a coating device (3) onto a first substrate (1), and a second substrate (4) is then disposed upon the free surface of the coated adhesive, by a nip roll (5). It is to be understood that this arrangement can be modified in other embodiments and especially that the second substrate  
25 (4) need not be used in all cases. Then, the nip roll (5) can be employed to nip the thermoplastic composition directly to the first substrate. For such embodiments, the nip roll (5) will be release-coated, e.g. may be a steel roller with a polytetrafluorethylene surface layer.

30 More specifically shown in Fig. 1A and 1B, Substrate 1 (1) travels past a series of idle rollers (2) to ensure the web is in proper alignment prior to approaching the coating device (3). Substrate 2 (4) is optionally adhered to the  
35 coating surface by means of a nip roll (5). Substrate 1 is defined as the first substrate that is contacted with the substantially continuous thermoplastic film. Substrate 1 may be any substrate which is generally provided in a roll good such as nonwoven, paper including release-coated paper,

5 and a wide variety of films, foils and other materials. The embodiment of Fig. 1A, where the nip roll (5) is located fairly remote from the contact point of adhesive film and first substrate, is especially suited for the coating of porous substrates. The embodiment of Fig. 1B is especially  
10 suitable when Substrate 1 is nonporous meaning air does not readily pass through the substrate. In the case of film lamination, Substrate 1 is typically a film. Substrate 2 may also be provided in a roll good and be the same or a different material as Substrate 1. However, Substrate 2 may  
15 also be a particulate substance such as superabsorbent polymer, or a release-coated web material that can be pulled off the adhesive coating.

Fig 1C shows an embodiment where the adhesive film is first  
20 nipped onto the first substrate (1) by nipp roll (5), which is part of a nipping station as later shown by rolls A and B in Figs. 2-10.

A second substrate 4 is then disposed on the free surface  
25 not in contact with the first substrate (1), at a lamination station formed by folls C and D.

Figs. 2-10 illustrate various preferred embodiments of the present invention wherein an extruded thermoplastic  
30 composition such as a hot melt adhesive is applied to a first substrate and then laminated to a second substrate. In each of these illustrations, Substrate 2 is optional in that the invention in its broadest aspect is simply a single continuous nonporous film formed from a non-contact coating  
35 method and coated onto a single substrate. In the absence of the second substrate, Figure 5B represents a transfer coat application since the molten composition is first applied to a release coated roller which then contacts a first substrate at the nip.



5 In embodiments where the thermoplastic coating or hot melt adhesive is contacted to a first substrate in the absence of a second substrate, as illustrated in Figures 6 and 7, or in the case when the second substrate is porous, it is important to have a release coating such as silicon, Teflon,  
10 or release paper on the roller(s) in contact with the adhesive or porous substrate to prevent adherence of the thermoplastic composition to the roller. The nip roller presses the air out from between the thermoplastic coating film and the substrate to insure there is no air entrapment  
15 between the first substrate and the thermoplastic composition. Roller A can be a steel cylinder to encourage heat transfer whereas roller B, typically the nip roller is rubber. In some cases it can be more preferred that roller A is rubber whereas roller B is a steel cylinder with an  
20 external release-coating.

Figs. 3-10 demonstrate that the nozzle position may be varied from perpendicular positions to parallel positions with respect to the position of the substrate.

25 Figs. 8 and 9 illustrate a second substrate being laminated to the first substrate at a position farther from the coating device. In this embodiment, it is preferred that roller C be heated to reactivate or extend the open time of  
30 the hot melt adhesive or thermoplastic coating prior to being laminated to the second substrate. The temperature of roller C can vary between about 30-100°C for lamination between rollers C and D. Alternatively, roller C may be a chill roll to hasten the speed of set of the thermoplastic  
35 coating or hot melt adhesive. This can be useful where the laminate is produced for intermediate storage. The substrate laminated in the nip of rollers can be either in web form, or in the form of sheets. As shown in Fig. 10, where roller C is a chill roll, the inventive method can be used to

5 produce substrates such as films coated on one side with a thermoplastic composition, which can e.g. be used for heat sealing applications. Where this is desired, a further layer of a release paper can of course be added, as shown in Fig. 9, to protect the heat-sealing material e.g. for  
10 intermediate storage.

The coating device is positioned at a distance of at least 0.5 mm, preferably at least 2 mm, from the substrate (or the release coated roller in the case of transfer coating in the  
15 absence of a second substrate - Fig. 5B). The maximum distance the coating device may be positioned from the substrate is only limited by practicality, particularly when the coating device is positioned substantially vertically. Preferably, the distance is less than about 5 m, preferably  
20 less than about 3 m, more preferably less than about 1 m, even more preferably less than about 500 mm, and most preferably from about 2 to 20 mm, depending on the properties of the thermoplastic composition being coating. It is typically advantageous that the area between the  
25 coating device and substrate be shielded during coating from air-borne contaminants and air currents to prevent distortion of the coating prior to contacting the substrate. This is particularly the case when the distance between the coating device and substrate is greater than about 500 mm.

30 The distance is largely dictated by the viscosity and open time of the thermoplastic composition being coated. In the case of producing barrier films in this manner, it is surmised that the thermoplastic composition cools  
35 sufficiently in its suspended state such that it has built in viscosity and cohesive strength to the extent that any filaments or fibers present on the substrate surface cannot penetrate the coating, yet the thermoplastic composition is molten enough to adequately adhere to the substrate. The

5 greater the distance between the coating device and the nip roller, the more the hot melt adhesive or coating will cool prior to contacting the first substrate. For some adhesive compositions, this cooling will adversely affect the adhesion (or anchorage) to the substrate. Therefore, the  
10 substrate may be passed over a heated roller prior to being nipped, or a heated nip roller may be employed if the distance between the nip roller and the coating device causes the coating or adhesive to cool to the extent that it will no longer adequately adhere or anchor to the substrate.

15

The coating may contact the substrate at any angle (compare e.g. Fig. 3 and 4). However, it has been shown to be especially advantageous for some applications such as for  
barrier films, that the coating later contacts the substrate  
20 in a substantially horizontal direction as in Fig. 1A, 1B, 2, 6 and 8. To accomplish this, a roller can be provided in the path of movement of the substrate to give the substrate a substantially vertical, upward direction, as the substrate passes the coating device. Additionally, the coating  
25 device, such as a slot nozzle, can be provided substantially horizontally beside the roller so that the coating travels from the side towards the surface of the substrate.

The diameter of the coating roller is preferably about 15 mm  
30 to about 50 mm in diameter with the nozzle slightly above the center of the coating roll such that the angle at which the thermoplastic coating contacts the substrate is less than about 60° as the substrate is moving away from the nozzle. The coating head is adjusted by one of ordinary  
35 skill in the art to optimize for even flow and distribution of the thermoplastic coating over the entire width of the application.

Thereafter, the sufficiently cooled coating contacts the

- 5 substrate surface and adheres to the surface without deeply penetrating into the substrate. If the thermoplastic coating is of such a composition that it substantially detackifies after sufficient cooling, the laminate of the coated substrate, thus formed, can be rolled up and stored.
- 10 Alternatively this can be achieved by placing a release coated second substrate, such as a silicone-coated paper, on the surface of the adhesive coating. The laminate can then be used at some later time. The laminate can be bonded by any suitable bonding technique including ultrasonic bonding,
- 15 heat sealing, or more commonly adhesive bonding.

Preferably, the coating is done "inline" immediately before any further processing. An example of an in-line process for which the invention is particularly well suited may be found in DE 195 46 272 C1 to Billhöfer Maschinenfabrik GmbH, incorporated herein by reference. The surface of the coating layer which is pointing away from the substrate may be sufficiently tacky such that it can be used as a construction adhesive or for lamination to other substrates and therefore can also serve to bond the coated substrate to another substrate layer. Other substrates that may be simultaneously bonded or laminated in this manner include absorbent, superabsorbent polymer, elastomeric strands or webs, tissue, films, foils, paper, cardboard, metal, as well as various permeable coverstock materials such as nonwoven or perforated films. These materials may be in the form of roll-goods, sheets, or particles.

In a preferred embodiment, the substrate to be laminated is paper or cardboard, especially printed paper, processed photographic paper or printed cardboard, as used in the production of e.g. book covers, picture postcards, calendars, posters, high quality packaging materials, gift-wraps and so forth. The laminating material can be

5 synthetic film material, paper, textile material or any other flexible laminating material suitable for lamination. Preferably, the laminating material is, however, a synthetic film material, especially a clear and transparent film material as is customarily used for such laminations.

10

Typically such film materials comprise plane or embossed films, which are at least substantially made from oriented polypropylene, polyethylene, polyesters such as Mylar®, polyacetate, nylon, celluloseacetate, and so forth having a  
15 thickness of about 5 microns to about 50 microns. These films are commonly laminated or sealed to printed paper or boardstock. Composite materials are commonly produced including film to film and film to foil and metallized substrates are commonly used for laminates. These types of  
20 laminates are commonly found in such industries as graphic arts and packaging. Using the method of the invention, such laminates can be produced using nonreactive hot melt adhesives instead of the commonly used reactive adhesives.

25 Generally, the exit temperature of the thermoplastic composition will be less than about 240°C, and thus much lower than typical polymer extrusion temperatures, which are of the order of 300°C. Although the temperature of the thermoplastic composition as it exits the coating device may  
30 range from about 80°C and about 180°C or more, the non-contact coating system of the present invention allows coating to be accomplished at extremely low temperatures. For this embodiment it is preferred that the thermoplastic composition be coated at a temperature less than 160°C, more  
35 preferably less than about 140°C, even more preferable less than about 120°C and even more preferable less than about 110°C. As mentioned previously, heat sensitive materials can also be coated in this manner by employing higher coating temperatures in combination with increasing the

5 distance between the coating device and the substrate to be coated to allow for sufficient cooling. Materials which are normally too sensitive mechanically and/or thermally (e.g. very low gauge films) for customary coating methods can therefore be coated using the method of the present  
10 invention. Such sensitive materials include low gauge polyethylene materials, low basis weight nonwovens and the like.

A substantial advantage of the present invention is that a  
15 substantially continuous coating layers can be made from hot melts at very low coating weights. Even with customary commercially available hot melts, continuous layers can be produced at coating weights ranging from about  $0.5 \text{ g/m}^2$  to as much as  $50\text{-}60 \text{ g/m}^2$ , preferably at coating weights of not  
20 more than about  $30 \text{ g/m}^2$ , more preferably at coating weights of not more than  $20 \text{ g/m}^2$ , even more preferably between  $10 \text{ g/m}^2$  and  $20 \text{ g/m}^2$  and most preferably less than  $10 \text{ g/m}^2$ . However, coating weights higher than  $60 \text{ g/m}^2$  may be useful for other applications wherein reducing the mechanical and  
25 heat-induced stresses is of primary importance.

The very thin coatings which can be produced according to the invention not only contribute to the economical advantages of the inventive method, but also makes it  
30 possible to achieve a very much reduced stiffness of the material, which thus comes much closer, in its properties, to uncoated substrates.

35 The thermoplastic composition

As previously mentioned various thermoplastic materials may be used in the present invention such as various thermoplastic polymers may be used including polyethylene,

5 polypropylene, copolymers of olefins, especially ethylene,  
and (meth-) acrylic acid; copolymers of olefins, especially  
ethylene, and (meth-) acrylic acid derivatives, especially  
(meth-) acrylic acid esters; copolymers of olefins,  
especially ethylene, and vinylic compounds, especially vinyl  
10 carboxylates such as vinyl acetate; thermoplastic rubbers  
(or synthetic rubbers) such as styrene-isoprene-styrene,  
styrene-butadiene-styrene, styrene-ethylene/butylene-styrene  
and styrene-ethylene/propylene-styrene block copolymers  
available in commerce under the tradenames of Kraton®,  
15 Solprene®, and Stereon®; metallocene-catalyzed polymers,  
especially based on ethylene and/or propylene; polyolefins  
such as ethylene, polypropylene and amorphous polyolefins  
(atactic poly- $\alpha$ -olefins) such as Vestoplast® 703  
(Hüls); polyesters; polyamides; ionomers and corresponding  
20 copolymers; and mixtures thereof. Such thermoplastic  
materials may be employed in the coating method of the  
present invention uncompounded provided the thermoplastic  
material is sufficiently low enough in viscosity. However,  
hot melt adhesives are preferred due to the ability to  
25 independently tailor the viscoelastic properties, open time,  
tack, and various other properties. Hot melt adhesives  
commonly have melt flow indices required for such processing  
already at very low temperatures. Typical hot melts are  
fluid enough for such processing at temperatures ranging  
30 from about 60°C to about 175°C. Additionally, various known  
hot melt moisture cure compositions are contemplated for use  
in the present invention.

With suitable hot melts, such as those described in DE-A-41  
35 21 716, it is also possible to make materials which are  
impermeable to liquid water, yet water vapor permeable  
rendering the coating "breathable".

5 In addition to commonly known hot melt adhesives, thermoplastic compositions comprising a water soluble, saline (body fluid) insoluble polymer such as Eastman AQ copolyesters, commercially available from Eastman, are also particularly useful for creating barrier films that are  
10 impervious to body fluid, yet readily water soluble. This feature is of particular interest for creating flushable and compostable disposable hygienic products. Furthermore, there may be applications wherein water permeability is desired. Accordingly, this coating method may also be  
15 suitable for coating water soluble and/or biodegradable thermoplastic materials.

In the case of the lamination adhesives for transparent substrates, thermoplastic polymers comprising substantially  
20 or consisting entirely of one or more ethylene/methylacrylate copolymers (EMA's) and/or ethylene/n-butyl acrylate copolymers (EnBA's) is preferred. EnBA copolymers are presently the most preferred such polymers.

25 More preferably, the thermoplastic composition exhibits certain rheological characteristics such that a substantially continuous coating can be produced at coating weights of less than about 50-60 g/m<sup>2</sup> and preferably less  
30 than about 30 g/m<sup>2</sup>. In general, the rheological properties preferably fall within a rheological window wherein the complex viscosity at the coating temperature at high shear rates (1,000 rad/sec) is less than about 500 poise and the complex viscosity at low shear rates (< 1 rad/sec) is less  
35 than about 1,000 poise. In other words, preferable thermoplastic compositions exhibit Newtonian regions at low shear rates and shear thinning at higher shear rates. Thermoplastic compositions having wide windows of application are those in which the composition exhibits the



5 appropriate rheological properties at a variety of application settings, particularly low application temperatures. Narrow application windows are those in which the rheological parameters are only met under very specific conditions.

10

The applicants surmise the complex viscosity and high shear relates to the processing conditions at the slot die exit. A composition with too high of a complex viscosity at 1,000 radians/sec would require excessive pump pressure to exit  
15 the coating device. A die with a shim gap larger than 3 mm could be used to process these materials but a higher coating weight may result.

The complex viscosity and low shear relates to the settling  
20 of the coating on the substrate during the time it is suspended above the substrate. If the low shear value is too high, the coating may not adhere adequately to the substrate and/or the thermoplastic composition builds up at the nozzle causing a streaked, discontinuous coating. If  
25 the low shear viscosity is too low, the coating may seep into the substrate, causing poor barrier properties.

Extensional viscosity, which was not measured can also strongly influence the melt strength. Higher levels of  
30 branching or the addition of a small concentration of a high molecular weight material can strongly influence the melt strength. More preferred, are compositions that meet the target rheological parameters at low application temperatures, less than about 177°C, preferably less than  
35 about 160°C, more preferably less than about 140°C, even more preferably less than about 125°C, most preferably less than about 110°C.

5 Accordingly, many known hot melt adhesive compositions are well suited for use in the coating method of this invention. Hot melt adhesives typically comprise at least one thermoplastic polymer, at least one plasticizer and at least one tackifying resin. Preferably, such suitable hot melts  
10 comprise up to 50% by weight of thermoplastic polymer, up to 40% by weight of a plasticizer and up to 70% by weight of tackifying resin. In the case of hot melt adhesives which are not pressure sensitive, wax is generally employed in concentrations up to about 30% by weight of the adhesive.

15

Generally, the invention's hot melts will additionally contain one or more tackifying resins, plasticizers or oils and waxes plus customary additives and adjuncts such as stabilizers, antioxidants, pigments, UV stabilizers or  
20 absorbers, fillers etc. Plasticizers and tackifying resins used in hot melt adhesives are known.

Oils such as naphthenic oils are preferred plasticizers. As for tackifying resins, those resins already known for such  
25 purposes are generally suitable, especially aliphatic, cycloaliphatic and/or aromatic hydrocarbon resins, ester resins and other such compatible resins. It is presently preferred to use either aliphatic or aromatic modified hydrocarbons resin. The preferred aliphatic resins are  
30 hydrogenated aliphatic hydrocarbon resin, for example, the Escorex® 5000 series available from the Exxon Chemical Co. in Houston, TX and the Arkon® P and M series available from Arakawa Chemical Co. and the Regalite® series available from Hercules Inc. in Wilmington, DE. Rosins and rosin  
35 ester resins are also useful in the present invention. One such hydrogenated rosin acid tackifying resin is Foral® AX available from Hercules. Modified hydrocarbon resins such as modified terpenes including styrenated terpenes such as

5 the Zonatac® series available from Arizona Chemical Co. in  
Panama City, FL and the Kristalex® series of alpha-methyl  
styrene resins available from Hercules, Inc. and the  
Uratack® series available from Arizona Chemical are also  
useful in the present invention. The components are mixed  
10 and processed in a known manner to prepare the hot melts  
which can be used according to this invention.

Waxes are also useful in the present invention. These  
include synthetic high melting point waxes such as Fischer  
15 Tropisch waxes available from Sasol (South Africa) under the  
tradename of Paraflint®, or from Shell Malaysia under the  
tradename Petrolite, and high density low molecular weight  
polyethylene waxes available from Marcus Chemical Co. under  
the tradename of Marcus®. AC 8 is another useful  
20 polyethylene wax available from Allied Chemical.  
Microcrystalline waxes and paraffin waxes are also useful to  
the present invention.

Laminating adhesives will preferably comprise up to 100% of  
25 at least one thermoplastic polymer described above; 0-50% of  
an aliphatic hydrocarbon resin; 0-20% of an aromatic  
hydrocarbon resin; 0-40% rosin and 0-20% wax, said  
components and their amounts being chosen so that the  
adhesive is in-line coatable onto a laminating material  
30 and/or a laminating substrate, for subsequent in-line  
lamination of said laminating material to said substrate.

More preferably, in the case of film laminating, the  
adhesive will comprise the following components: up to 100%  
35 of at least one EMA and/or EnBA copolymer; 0-50%  
hydrogenated aliphatic hydrocarbon resin; 0-20% alpha-methyl  
styrene resin; 0-40% hydrogenated rosin and 0-20%  
polyethylene wax.

- 5 The hot melt adhesive usable for practicing the invention's method can, in the simplest case, consist substantially or even completely of one or more grades of EMA or EnBA copolymers. EMA and EnBA copolymers are available from Elf Atochem under the Lotryl® tradename, from Quantum Chemical
- 10 Co. and From Exxon Chemical Co. under the Optema® tradename. A variety of different grades of EMA and EnBA copolymers are available. They mainly differ in ester content, melt flow index (MFI) and melting point.
- 15 In presently preferred special embodiments, the hot melt adhesive essentially consists of 35-60% EnBA or EMA; 30-50% hydrogenated aliphatic hydrocarbon resin or about 10% alpha-methyl styrene resin; 0-30% hydrogenated rosin and 0-10% polyethylene wax, plus small amounts of stabilizer. In some
- 20 preferred embodiments, the thermoplastic polymer of the hot melt adhesive is a single grade of EnBA copolymer, usually at the low end of the MFI range (i.e. MFI less than 10 g/10 min.) In other preferred embodiments, the thermoplastic polymer comprises more than one grade of EnBA, and in these
- 25 cases, two or three different grades wherein at least two of the grades preferably have MFI's which differ by at least a factor of 4 and up to a factor of 10 (i.e. one grade has an MFI more than 4 times that of the other grade.
- 30 The inventive hot melts can be used at application temperatures (or processing temperatures) which are low enough to prevent distortion of heat sensitive plastic film, and at the same time show excellent flow properties at such low temperature. It is, for example, possible to coat and
- 35 laminate the inventive hot melt on the laminating materials. Non-contact coating is especially advantageous for heat sensitive films. Excellent film forming performance is achieved, and the laminated products exhibits high glossiness.

5

The laminating adhesives of the invention produce high transparency of the hot melt coating, so that high gloss is achieved, while readability and color rendition of, for example, printing on the substrate is not impaired.

10

The inventive hot melts show excellent (high) hot-tack and open time characteristics for the method of the present invention as well as setting properties. They meet the requirements of machine condition, in-line embossing and

15 cutting, for example, in the graphic arts industry.

Laminates made according to the invention exhibit high heat resistance and high UV resistance, and correspondingly little delamination or yellowing. Also after heat forming

20 and embossing, no delamination is observed when the hot melt formulations of the invention are used.

The following non-limiting examples further assist in illustrating the present invention.

25

#### Examples

Hot melt adhesives were produced from different

30 thermoplastic polymers, tackifiers and plastizisers as shown in Table 1 below:

Examples 1-8

TABLE I

Ingredients	Ex 1	Ex 2	Ex 3	Ex 4	Ex 5	Ex 6	Ex 7	Ex 8
Lotryl® 17 BA 07	23	40	35	10	23	-	-	-
EnBA copolymer								
Lotryl® 35 BA 40	15	-	-	20	15	20	15	15
EnBA copolymer								
Lotryl® 35 BA 320	17	-	-	30	17	10	16	15
EnBA copolymer								
Escorene® UL 150-19	-	-	-	-	-	20	24	23
EVA copolymer								
AC-8	5	10	-	-	5	-	5	-
polyethylene wax								
Parafint® C 80	-	-	-	-	-	10	-	-
polyethylene wax								
MOBIL Wax 145	-	-	-	-	-	-	-	5
paraffin wax								
Escorez® 5300	28	38	38	38	-	23	28	30
hydrocarbon resin								
Foral® AX	10	10	25	-	28	15	10	10
rosin acid resin								
Kristalex® F 85	-	-	-	-	10	-	-	-
alpha-methyl styrene resin								

5 Hot melt adhesives corresponding to the compositions depicted in Examples 1 and 7 were coated onto substrates, using a modified PAK 600 laminating machine by Kroenert, Hamburg, Germany. The structure of this machine is basically similar to that shown in Fig. 1B. With this type  
10 of machine, it is possible to nip the adhesive film directly onto the first substrate (1) by means of nip roller (5) or nip a second substrate (4) onto the first substrate and adhesive, again by means of nip roller (5). In the tests, both methods were tried. The dispensing temperature of the  
15 hot melt was 140°C for the composition of Example 1, and 110°C for the composition of Example 7. These compositions show favourable low viscosities, as is notable from the diagram Fig. 14. This diagram illustrates the viscosities of Examples 1 and 7.

20 Coatings were made on polyester film (Polyester RN 36, produced by Pütz Folien, Taunusstein-Wehen, Germany) and high density polyethylene films (HDPE KC 3664.00, obtained from Mildenerger + Willing, Gronau, Germany).

25 As a second substrate (where used), these films were also used. In other experiments, silicone paper was used instead. Tests were also made with printing paper sheets as the second substrate.

30 Coating weights were 5 to 6 g/m<sup>2</sup> at machine speeds of approximately 70 m/minute.

The adhesive film was released from the coating slot nozzle,  
35 at various distances from the first substrate (1) to be coated with the adhesive, in a variety of tests. In another set of experiments, with a vertical configuration (similar to Figs. 3-5, 7, 9 and 10) it was found that the distance of the slot nozzle from the substrate could be varied between a

- 5 few millimeters and up to 500 mm and more, without materially affecting the quality of the coating.

Wherein these experiments, the adhesive film released from the coating slot nozzle was directly coated onto the first  
10 substrate by means of nip roller (5) provided with a release coating, it was found that the adhesive did not adhere to the nip roller. The nip pressure was not measured, but the nip roller was pressed against the substrate at a laminating pressure of 7 to 8 bar.

15

It was found that the adhesive coated onto the first substrate left the nip station with no air enclosed between the adhesive and the first substrate.

- 20 In other tests, a second substrate was laminated onto the adhesive layer by a second set of rollers, located in the flow path of the substrate upstream of the nip roller (5). Also these laminations, using the same films, or release-coated paper, as above discussed, were examined for  
25 streaking, enclosed air, or other lamination defects.

The laminations thus made were all free of flaws. No streaking, enclosed air or any other defects were observed.

- 30 In a similar fashion, laminations were made using the same type of films, but the other adhesives depicted in Examples 2 to 6 of Table 1. The results were as good as those obtained with the adhesive compositions of Examples 1 and 7.

35



5

C L A I M S

1. A method of coating, wherein a hot melt adhesive,  
10 which has been thermally made flowable, is released from a  
coating device onto a substantially nonporous substrate as  
a substantially continuous coating without contact between  
said coating device and said substrate, and subsequently  
disposed upon the surface of said substrate.

15

2. A method of coating, wherein a hot melt adhesive,  
which has been thermally made flowable, is released from a  
coating device onto a substrate as a substantially  
continuous coating without contact between said coating  
20 device and said substrate, and subsequently disposed upon  
the surface of said substrate, wherein the distance between  
the coating device and the substrate is greater than 20 mm.

3. A method of coating, wherein a hot melt adhesive,  
25 which has been thermally made flowable, is provided in the  
form of a substantially continuous nonporous film without  
contact of the film with a substrate or a roller, and said  
film is then coated onto a substrate by means of a release-  
coated roller in direct contact with the adhesive film,  
30 said roller nipping said adhesive and said substrate.

4. A method of coating, wherein a hot melt adhesive,  
which has been thermally made flowable, is released from a  
coating device onto a release coated roller as a  
35 substantially continuous coating without contact between  
said coating device and said roller, and subsequently  
disposed upon the surface of a substrate.

5. A method of coating, wherein a hot melt adhesive,

5 which has been thermally made flowable, is provided in the  
form of a substantially continuous nonporous film without  
contact of the film with a substrate, and said film is then  
coated onto a first substrate, with a release-coated  
substrate disposed upon that surface of the hot melt  
10 adhesive which is not in contact with the first substrate.

6. A method of coating, wherein a hot melt adhesive,  
which has been thermally made flowable, is provided in the  
form of a substantially continuous nonporous film without  
15 contact of the film with a substrate, and said film is then  
disposed upon a release-coated substrate and is then  
transfer-coated onto a second substrate.

7. A method of coating, wherein a hot melt adhesive,  
20 which has been thermally made flowable, is released from a  
coating device onto a first substrate as a substantially  
continuous coating without contact between said coating  
device and said first substrate, and subsequently disposed  
upon the surface of said first substrate, wherein said  
25 coating is reheated and then contacted to a second  
substrate.

8. A method of coating, wherein a thermoplastic  
material which has been thermally made flowable, is  
30 provided in the form of a substantially continuous  
nonporous film without contact of the film with a substrate  
and said film is then coated onto a substantially nonporous  
substrate.

35 9. A method of coating, wherein a thermoplastic  
material, which has been thermally made flowable, is  
released from a coating device at less than about 240°C in  
the form of a substantially continuous nonporous film,  
without contact of the film with a substrate, and said film

5 is then coated onto a substrate.

10. A method of coating, wherein a thermoplastic material, which has been thermally made flowable, is provided in the form of a substantially continuous  
10 nonporous film without contact of the film with a substrate and said film is then coated onto a substrate, said coating having a complex viscosity of less than about 500 poise at about 1000 radians/sec at the coating temperature.

15 11. A method of coating, wherein a thermoplastic coating having a complex viscosity of less than about 500 poise at about 1000 radians/sec at the coating temperature has been thermally made flowable is released from a coating device  
20 onto a nonporous substrate as a substantially continuous coating without contact between said coating device and said substrate, and subsequently disposed upon the surface of said substrate.

12. A method of coating, wherein a thermoplastic coating  
25 having a complex viscosity of less than about 500 poise at about 1000 radians/sec at the coating temperature, which has been thermally made flowable, is released from a coating device onto a substrate as a substantially continuous coating without contact between said coating  
30 device and said substrate, and subsequently disposed upon the surface of said substrate, wherein the distance between the coating device and the substrate is greater than 20 mm.

13. A method of coating, wherein a thermoplastic coating  
35 having a complex viscosity of less than about 500 poise at about 1000 radians/sec at the coating temperature, which has been thermally made flowable, is released from a coating device onto a first substrate as a substantially continuous coating without contact between said coating

5 device and said first substrate, and subsequently disposed upon the surface of said first substrate, wherein said coating is reheated and then contacted to a second substrate.

10 14. A method of coating, wherein a thermoplastic coating having a complex viscosity of less than about 500 poise at about 1000 radians/sec at the coating temperature, which has been thermally made flowable, is released from a coating device onto a release coated roller as a  
15 substantially continuous coating without contact between said coating device and said roller and subsequently disposed upon the surface of a substrate.

15. The method of claims 8 to 14, wherein said  
20 thermoplastic coating has a complex viscosity of less than 1000 poise at about 1 radians/sec at the coating temperature.

16. The method of claims 1 to 15, wherein said substrate  
25 is selected from the group consisting of film, foil, paper and combinations thereof.

17. The method of claim 16, wherein said first and said second substrates are selected from films, foils, paper,  
30 coated paper, co-extruded films and other laminating materials, and said adhesive is a nonreactive adhesive, or a reactive hot melt adhesive.

18. The method of claims 15 or 16, wherein said coated  
35 substrate comprises a heat sealing material.

19. The method of claims 1 to 18, wherein said coating device is a slot nozzle.

5       20. The method of claims 1 to 19, wherein said coating  
has an area weight of less than about 60 g/m<sup>2</sup>.

21. The method of claims 1 to 20, wherein the coating  
has an area weight of less than about 30 g/m<sup>2</sup>.

10

22. The method of claims 1 to 21, wherein the coating  
has an area weight of less than about 10 g/m<sup>2</sup>.

23. The method of claims 8 to 14, wherein the  
15 thermoplastic composition is released from the coating  
device at a temperature of less than about 160°C.

24. The method of claims 8 to 14, wherein the  
thermoplastic composition is released from the coating  
20 device at a temperature of less than about 110°C.

25. The method of claims 1 to 24, wherein a first  
substrate is bonded to at least one second substrate  
„inline“ or „offline“.

25

26. The method of claims 1 to 25, wherein the distance  
between the coating device and the substrate ranges from  
about 0.5 mm to 500 mm.

30       27. A book cover made by any one of the methods of claim  
1 to 26.

28. A lamination made by any one of the methods of  
claims 1  
35 to 26.

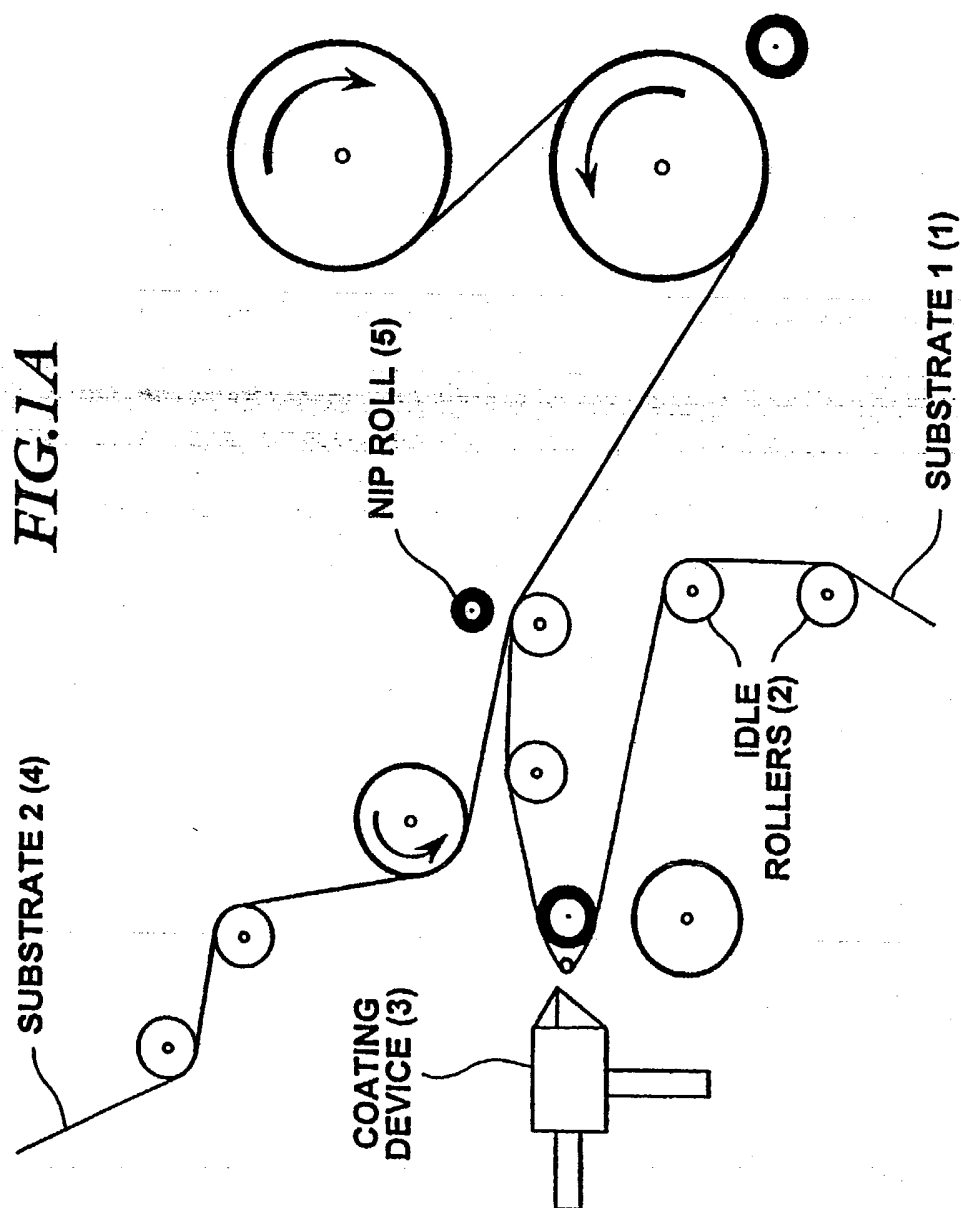


FIG. 1B

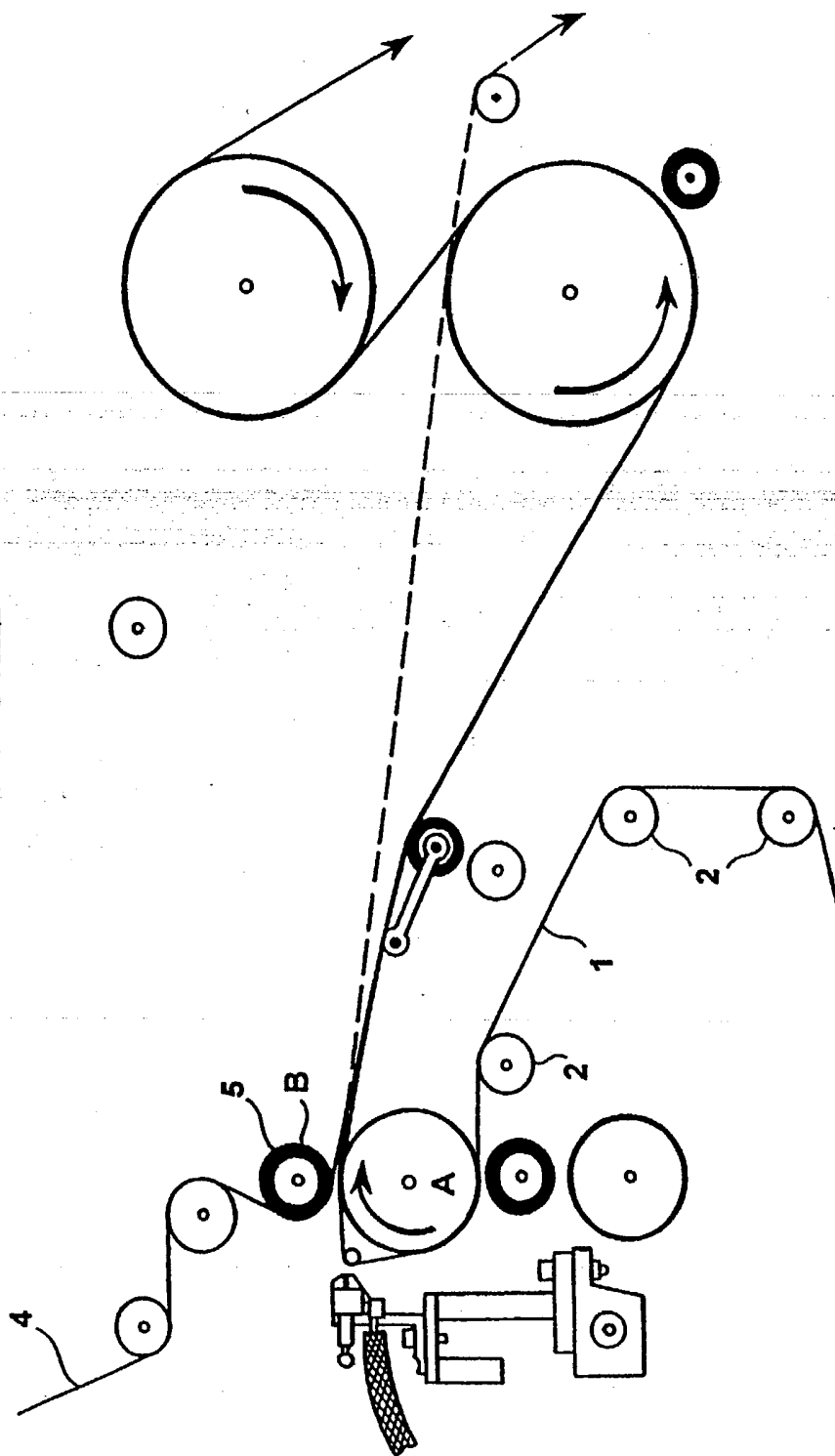
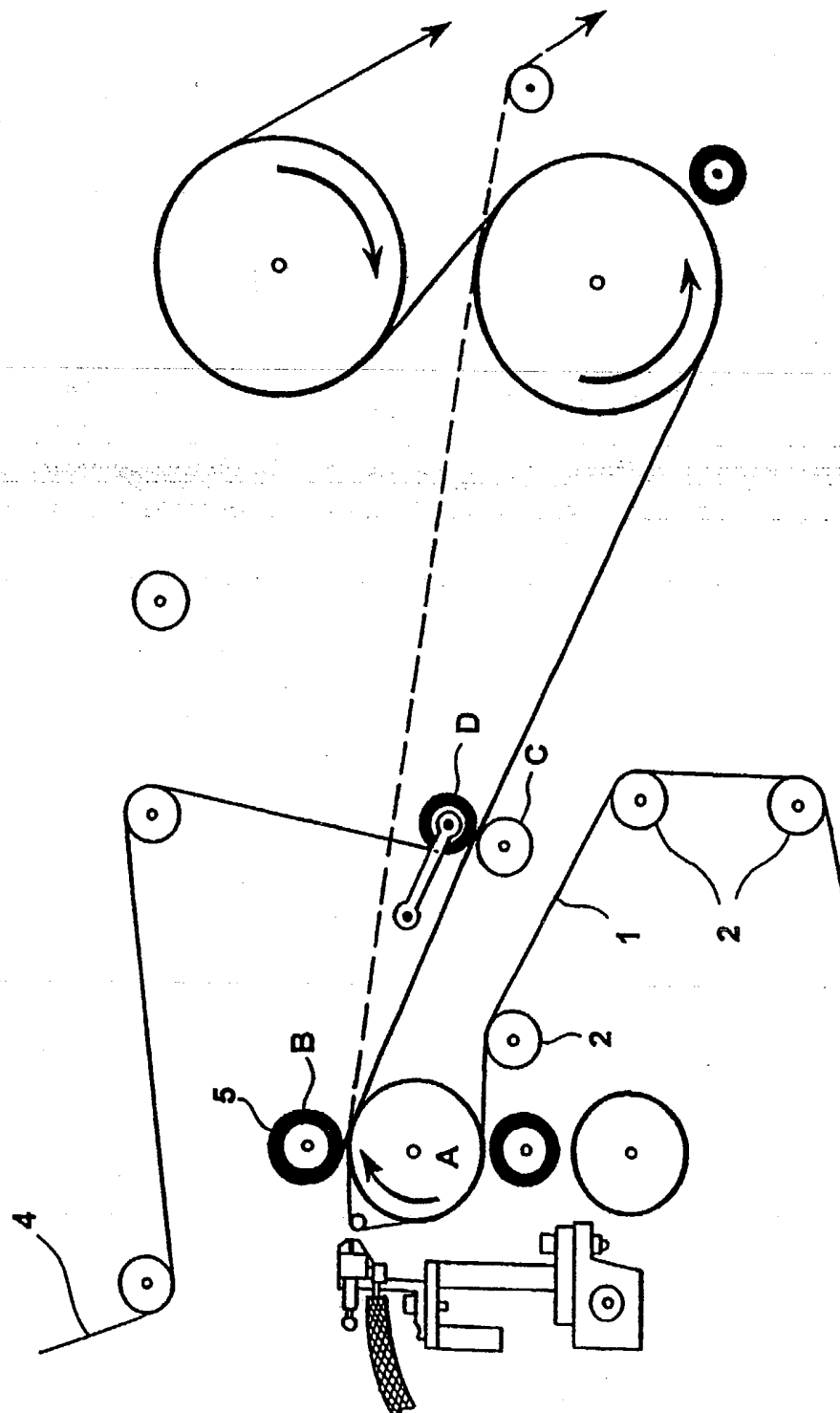
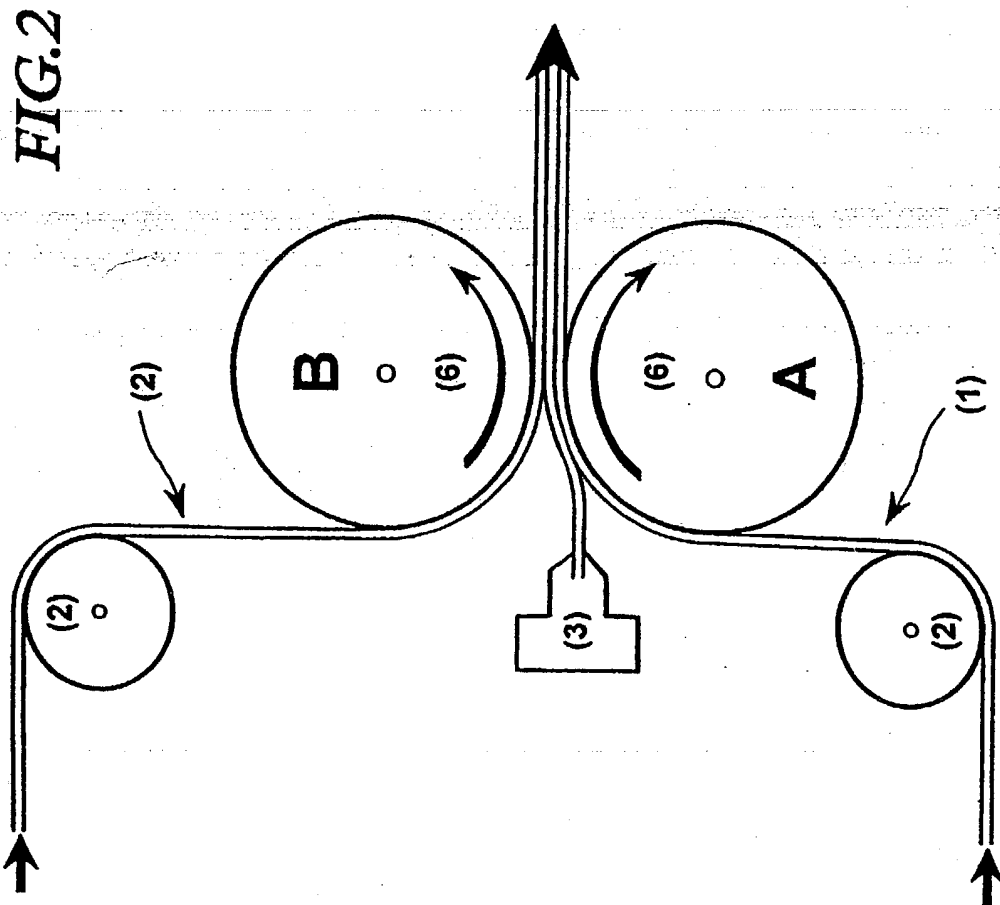


FIG.1C

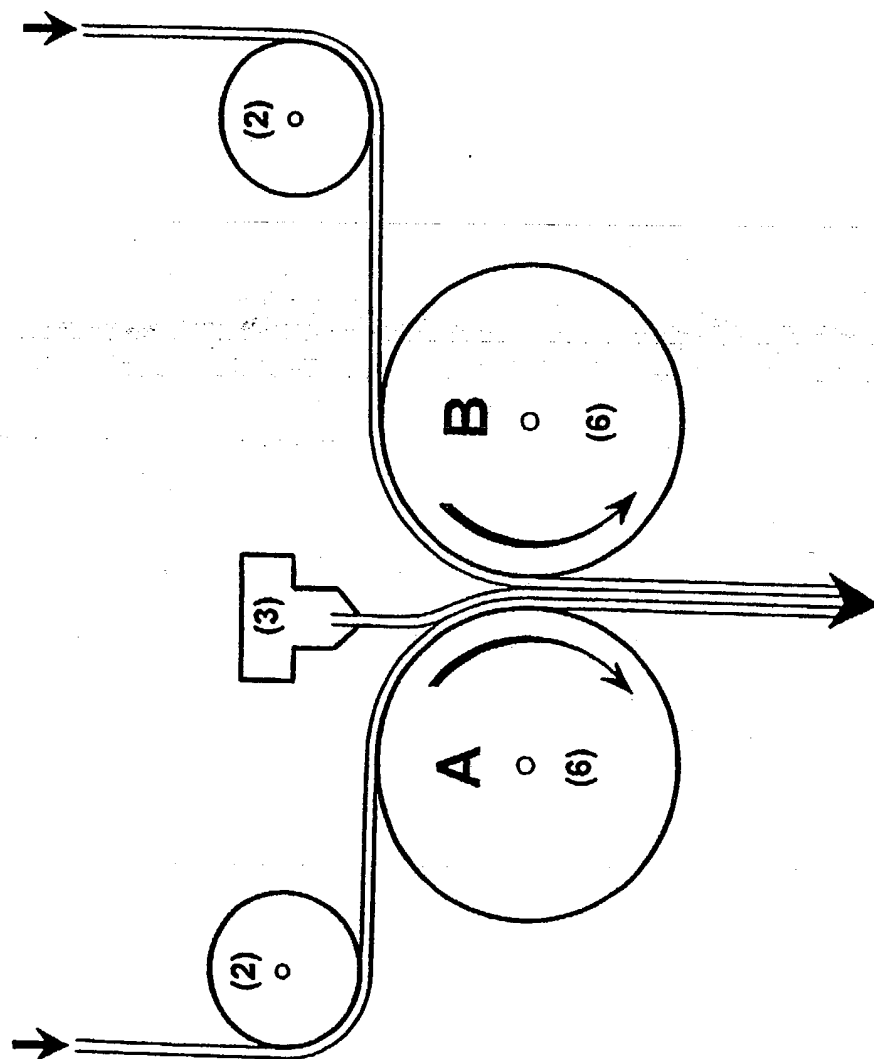






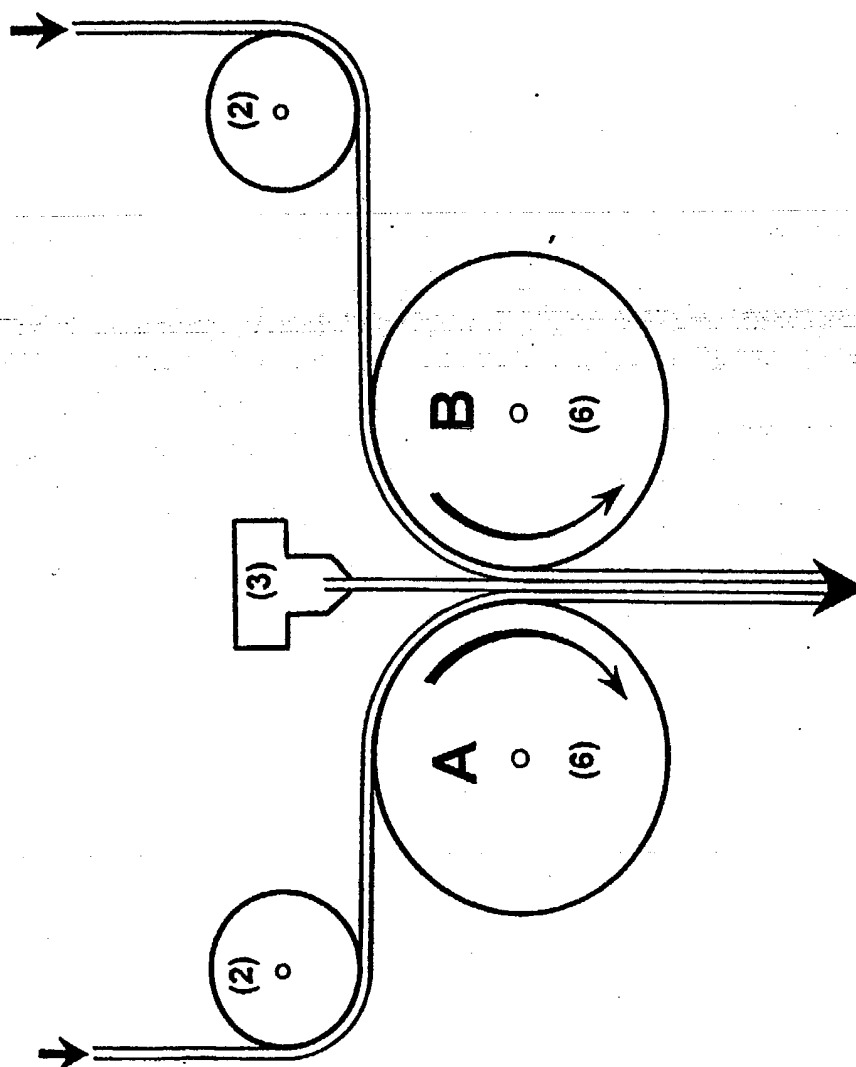
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FIG.3



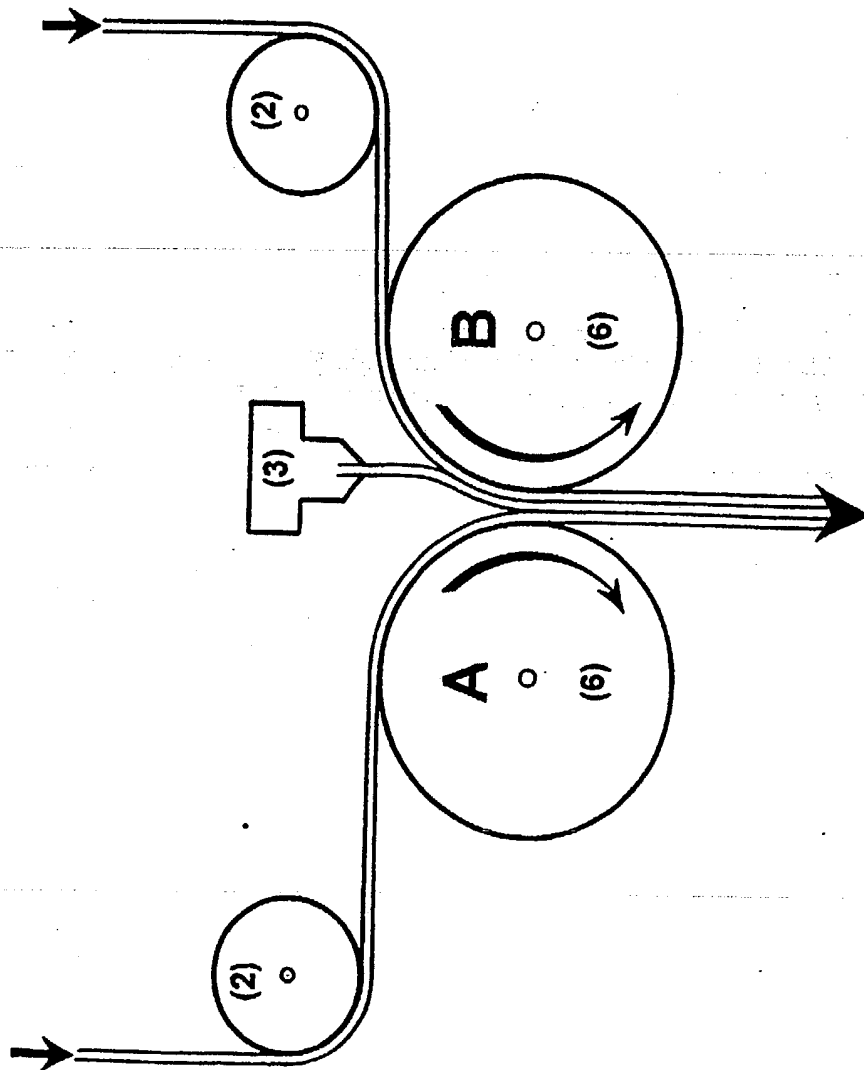
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FIG.4



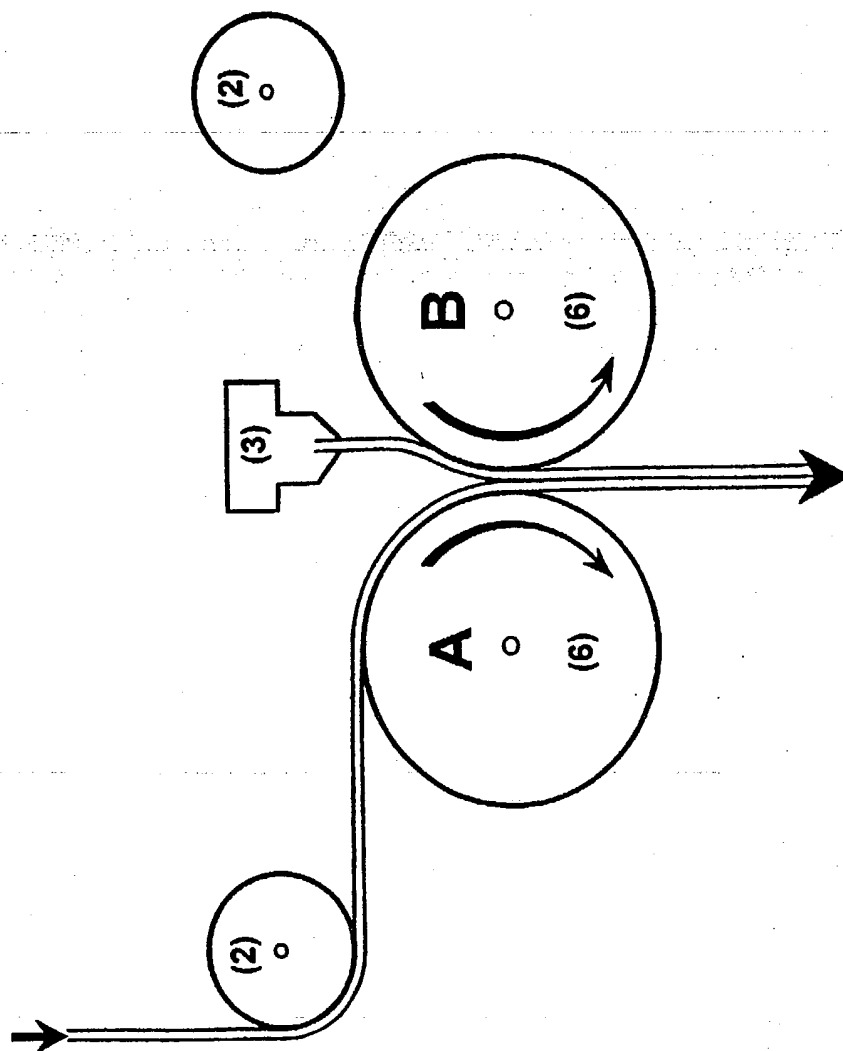
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FIG.5A



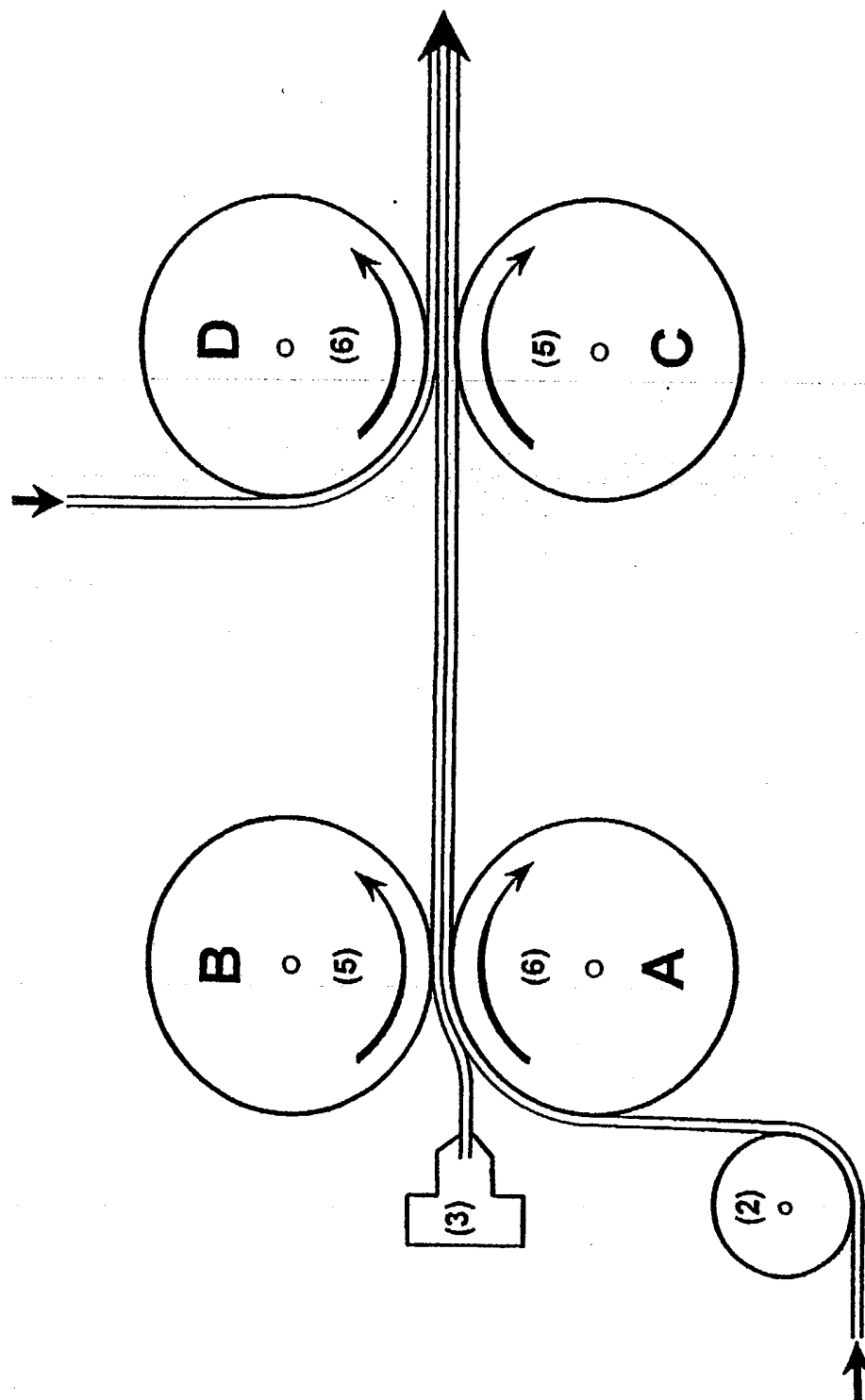
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FIG.5B



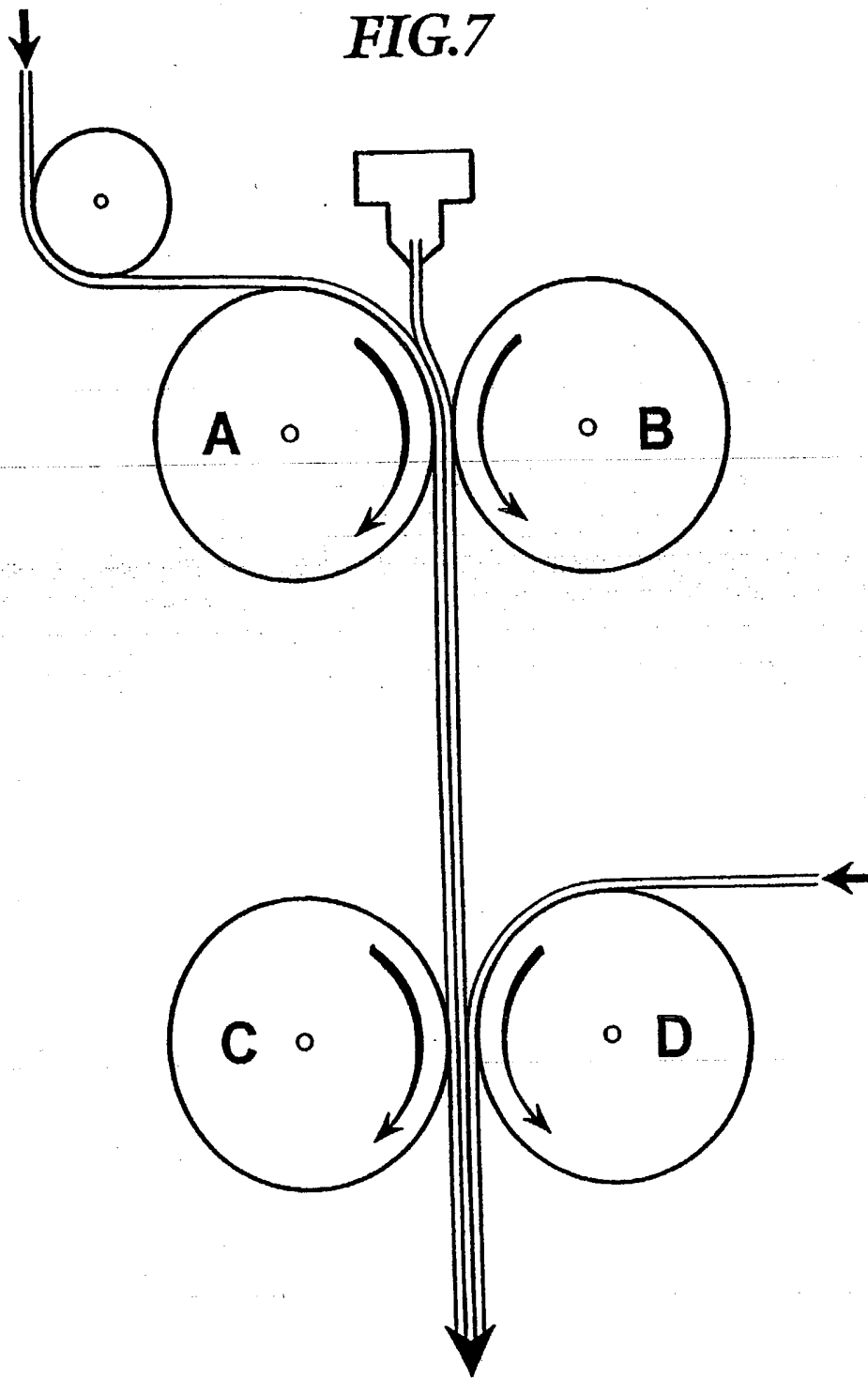
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FIG.6

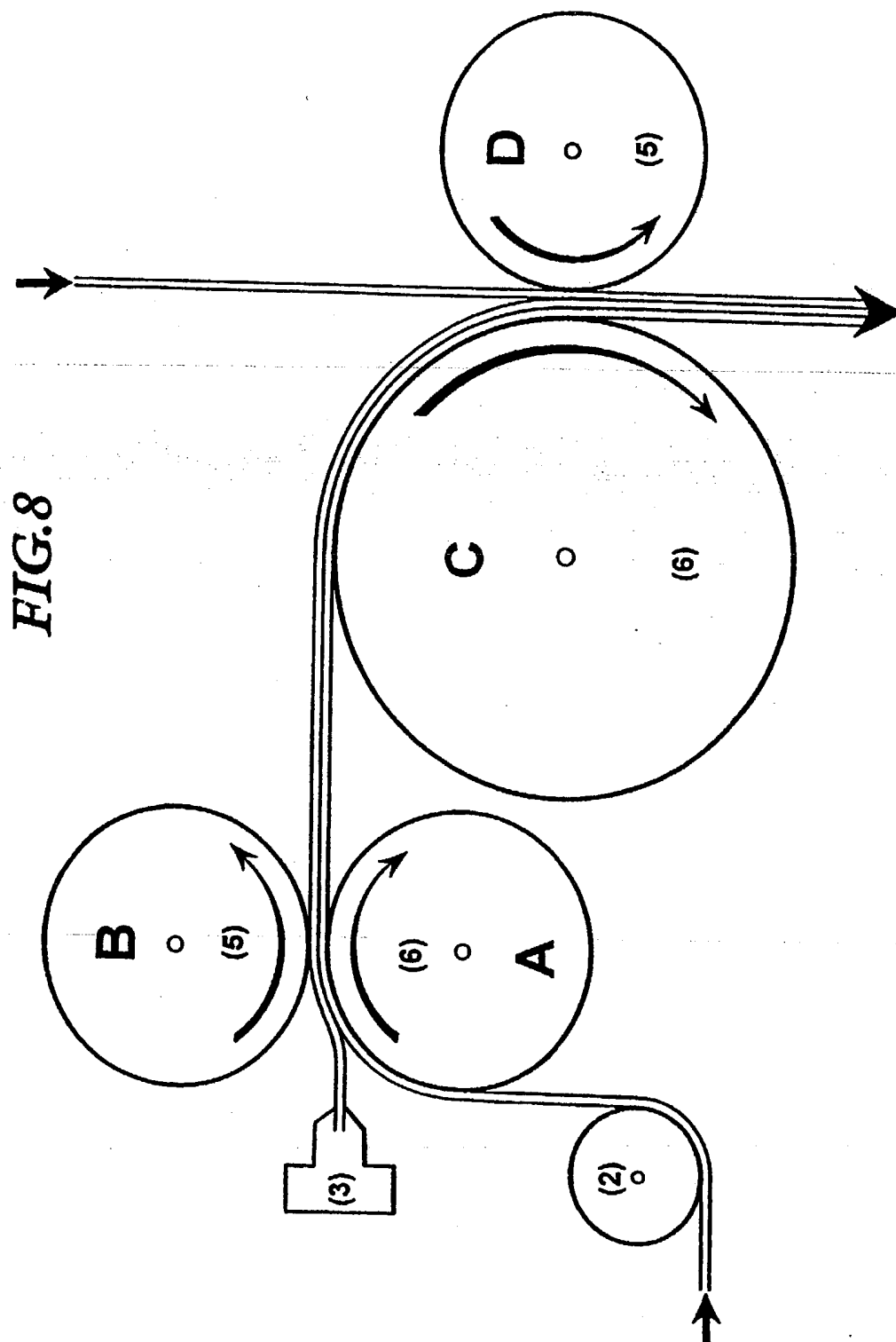


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**FIG.7**



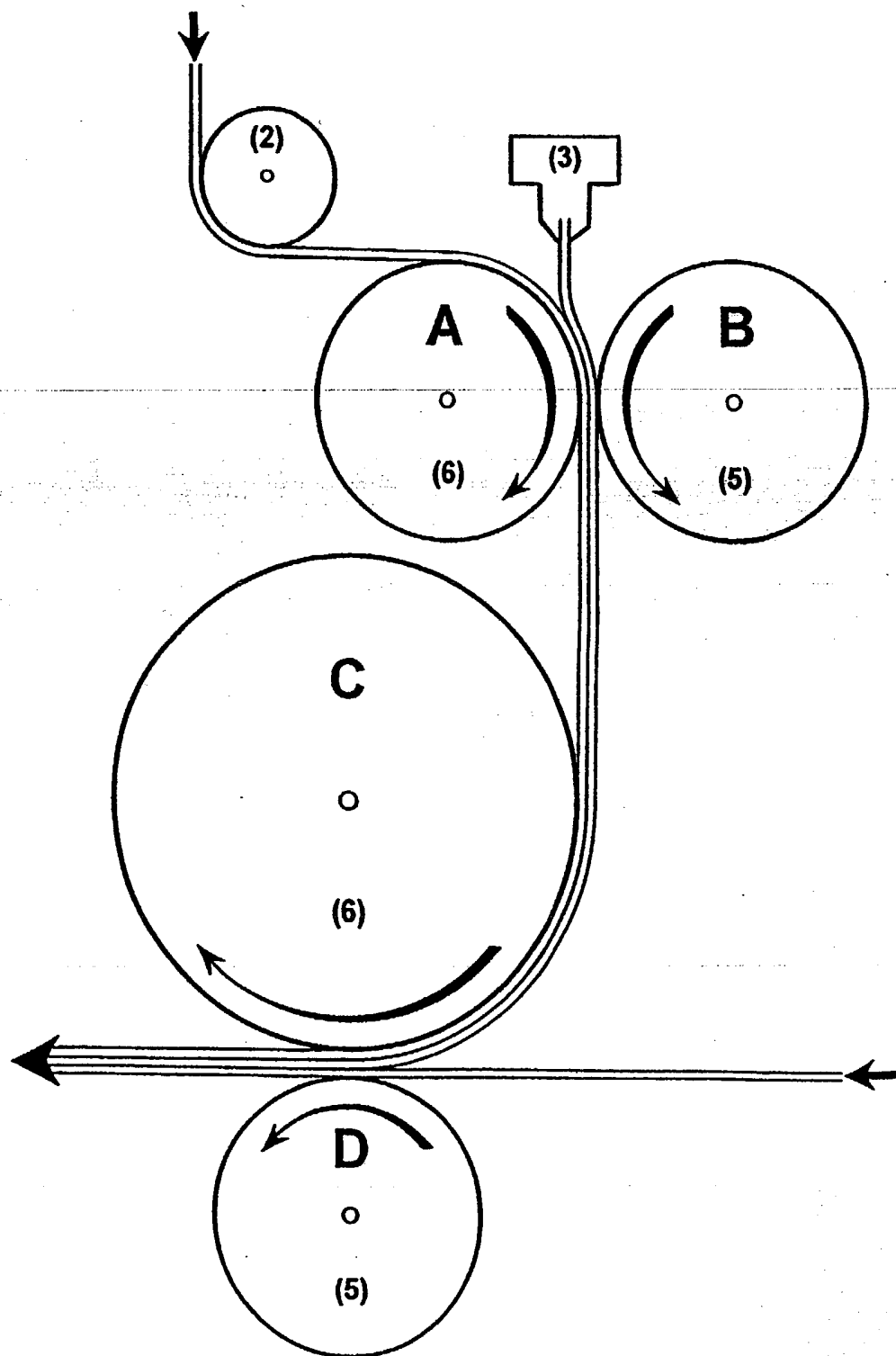
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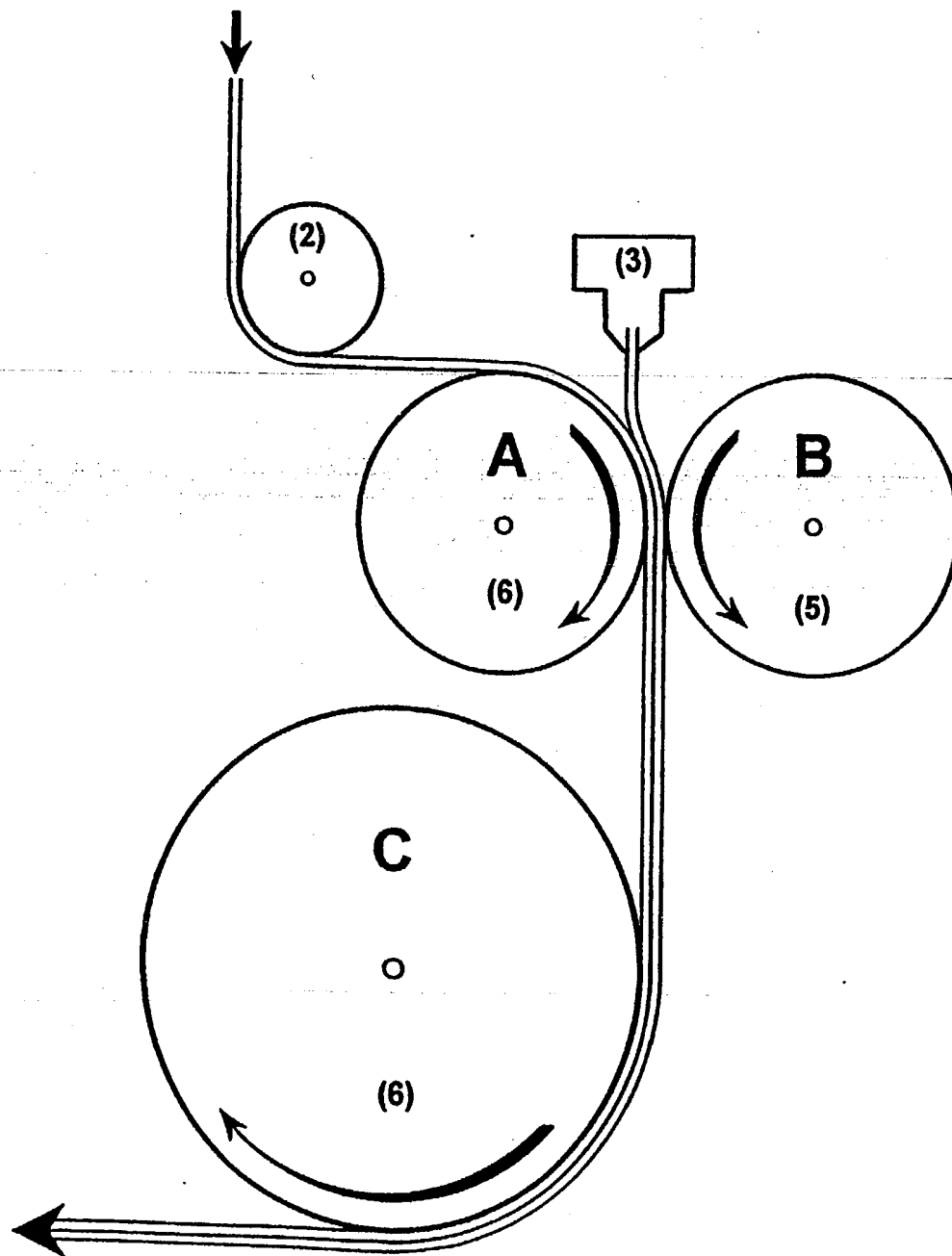
12/14

**FIG.9**



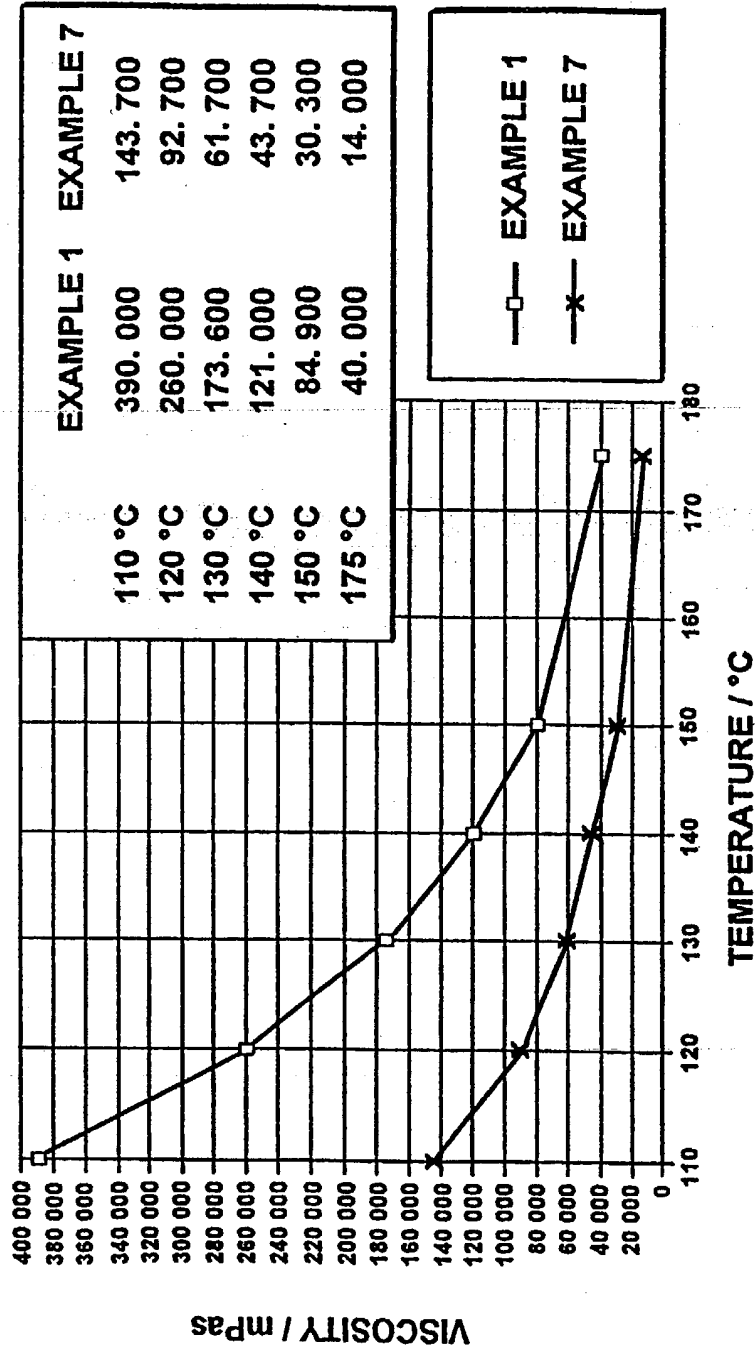
13/14

**FIG.10**



**FIG.14**

**VISCOSITY TEMPERATURE FUNCTION**



# INTERNATIONAL SEARCH REPORT

Int. l. Application No

PCT/EP 98/01588

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 B05D1/26 B05D5/10 B05D1/30 B32B31/00 C09J7/02

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 B05D B32B C09J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	GB 2 274 429 A (XEROX CORP) 27 July 1994  see the whole document	1-4,7,8, 16,17, 19,25,27
X	WO 97 15722 A (MINNESOTA MINING & MFG) 1 May 1997  see the whole document	1,3,4,9, 16, 19-21, 23,26
X	US 4 407 689 A (OHTSUKI AKIRA ET AL) 4 October 1983  see the whole document	1,8,9, 16,17, 19,23, 25,28

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☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax: (+31-70) 340-3016

Authorized officer

Brothier, J-A

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